Spin nematics and magnetization plateau transition in anisotropic Kagome magnets

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We study $S = 1$ kagome antiferromagnets with isotropic Heisenberg exchange J and strong easy axis single-ion anisotropy D. For $D \gg J$, the low-energy physics can be described by an effective $S = 1/2$ XXZ model with antiferromagnetic $J_z \sim J$ and ferromagnetic $J_\perp \sim J^2/D$. Exploiting this connection, we argue that non-trivial ordering into a "spin-nematic" occurs whenever D dominates over J, and discuss its experimental signatures. We also study a magnetic field induced transition to a magnetization plateau state at magnetization 1/3 which breaks lattice translation symmetry due to ordering of the S^z and occupies a lobe in the B/J_z-$J_z/J_\perp$ phase diagram.

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Introduction: Magnets with *geometrical frustration* (competition between different spin exchange interactions caused by the lattice geometry) exhibit many interesting properties including spin-liquid like low temperature phases and unusual spin correlations[1,2]. Kagome lattice (see Fig 1) magnets provide many examples of this, and several realizations with spins ranging from $S = 1/2$ to $S = 5/2$[3,4,5] have been experimentally studied. On the theoretical side, numerical and analytical work suggests that $S = 1/2$ isotropic Heisenberg antiferromagnet on the kagome lattice is in an unusual phase with an anomalously large density of singlet excitations[6,7] at $B = 0$. At finite B, there also exists evidence for the presence of a robust magnetization plateau state with magnetization pinned to 1/3 of the saturation moment[8].

A particularly interesting example of frustrated magnetism is provided by the Ni^{2+} based effective $S = 1$ Kagome magnet $Ni_3V_2O_8$, in which isotropic exchange interactions compete along with sizeable single-ion anisotropy terms (and weak Dzyloshinski-Moriya interactions) resulting in a rich phase diagram in the presence of a magnetic field[9,10]. Motivated in part by this, we consider a kagome lattice model with nearest neighbour antiferromagnetic spin exchange interaction ($J > 0$) between spin $S = 1$ ions in the presence of an easy-axis single-ion anisotropy ($D > 0$) along the z axis with the Hamiltonian

$$H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - D \sum_i (S_i^z)^2 - B \sum_i S_i^z , \quad (1)$$

where $\langle ij \rangle$ refer to nearest neighbour links of the two-dimensional kagome lattice.

Unlike the *unfrustrated case*, the D term may have important effects in frustrated systems even if not very big. We therefore study the limit of large D/J and show that interesting physics emerges: We show that the ground state at $B = 0$ is a quantum spin nematic associated with ordering of $< (S^+)^2 >$ without ordering of the spin itself. Upon increasing the field, magnetization plateaus appear at specific magnetization values. Of particular interest is a plateau at magnetization 1/3 which we show breaks translational symmetry. The corresponding plateau transition has a number of interesting properties which we discuss.

When D/J is large and positive and $B \lesssim J$, each spin is predominantly in the $m_z = \pm 1$ states, and we can describe the low energy physics in terms of an effective Hamiltonian for (pseudo-) spin $S = 1/2$ variables σ^z. Explicit calculation to second order in D/J yields the following effective low energy Hamiltonian in this regime[11]:

$$H_{\rm eff} = -\frac{J_\perp}{4} \sum_{\langle ij \rangle} (\sigma_i^+ \sigma_j^- + h.c.) + \frac{J_z}{4} \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - B \sum_i \sigma^z ,$$

Here, the $\vec{\sigma}$ are the usual pauli spin matrices, and the parameters of $H_{\rm eff}$ are given by $J_z \approx 4J + J^2/D$ and $J_\perp \approx J^2/D$; thus, for large D/J we have $J_z/J_\perp \approx 4D/J + 1 + O(J/D)$. Clearly, the ground-state of this pseudospin $S = 1/2$ XXZ model for small $J_z/J_\perp$ (which is not directly related to the physics of our original $S = 1$ problem) must be a ferromagnet polarized in the xy plane. Below we analyze the large $J_z/J_\perp$ regime (appropriate for the large D physics of the original $S = 1$ model) separately for $B = 0$ or small, and $B \sim J$.

When $B = 0$, the dominant diagonal interaction J_z leads to frustration since it is impossible to have all pairs of neighboring spins pointing anti-parallel to each other along the z axis on the kagome lattice. The ground state then lives entirely in the highly degenerate minimally frustrated subspace with precisely one frustrated bond (parallel spins) per triangle, and is selected by the spin-exchange dynamics ($J_\perp$). This physics in the present $J_\perp > 0$ case can be understood straightforwardly by thinking in terms of variational wavefunctions (as was done recently[12,13] on the triangular lattice): Since the spin-exchange $J_\perp > 0$ is *unfrustrated*, a good variational wavefunction for the small $J_z/J_\perp$ ferromagnet is simply $|\Psi_F\rangle = \Pi_i |\sigma_i^x = +1\rangle$. Furthermore, a natural description

for the state at *large* $J_z/J_\perp$ can be obtained by projecting $|\Psi_F\rangle$ to the minimally frustrated subspace described above. Since this subspace admits considerable fluctuations in the values of σ_z, such a projected wavefunction $|\Psi_\infty\rangle$ continues to gain 'kinetic energy' from spin-exchange processes, while minimizing the diagonal interaction energy by construction.

Thus, *x-y* ferromagnetic order *persists even in the large* $J_z/J_\perp$ *limit* at $B = 0$, and this remains valid for small B as well. Moreover, σ^z correlators in $|\Psi_\infty\rangle$ are simply given by the $T = 0$ correlations of the classical Ising model on the Kagome lattice, and their short-ranged nature[14] rules out any co-existing σ^z spin density wave order. [The same conclusion has been reached recently in other ways[15] and confirmed numerically[16].] What does this analysis imply for the original $S = 1$ magnet? As the pseudospin operator $\sigma^+ \sim (S^+)^2$, the xy ferromagnet of the pseudospin magnet actually corresponds to an *xy spin nematic* state where $< (S^+)^2 >\neq 0$ but $< \vec{S} >= 0$. Thus, we conclude that spin-1 Kagome magnets with strong easy axis anisotropy order into such a spin nematic phase with $< (S^+)^2 >\neq 0$ for $B = 0$ and its immediate vicinity.

The presence of this nematic ordering is one of our main conclusions. As a state that breaks the global $U(1)$ symmetry of spin rotations about the easy axis this nematic will have a gapless linear dispersing 'spin' wave which will lead to a T^2 contribution to the low temperature specific heat. Further this state will have a non-zero finite spin susceptibility for fields both parallel and perpendicular to the easy axis. Despite these similarities with conventional ordered antiferromagnets there will not be any magnetic Bragg spots in neutron scattering as the spin itself is disordered.

In passing we note that the same considerations on a triangular lattice again predict nematic ordering which *coexists* with spin density wave ordering of the z-component of the spin - this follows directly from the arguments above and the results of Ref. 12,13,17.

Returning to the Kagome lattice, as we turn on a magnetic field B, the magnetization will initially rise smoothly with field since the nematic persists for small B. As the field is increased to $B \sim J$, there will be plateaus where the magnetization is field independent and fixed to specific commensurate values of the magnetization. We now show that for a range of B away from $B = 0$ there is such a plateau state at magnetization 1/3 where the ground state is a lattice-symmetry broken spin-density wave (SDW) state (in which the z component of the spins order as in Fig 1).

Working again with the effective XXZ pseudospin Hamiltonian we begin in the extreme limit of $J_\perp/J_z \to 0$ by writing B in terms of a reduced field b as $B = J_z b$ and noting that the z coupling and field terms in $H_{\rm eff}$ can be combined and rewritten as $\frac{J_z}{8}\sum_t(\sigma^z_t - 2b)^2$, where the sum is now over all triangles t of the kagome lattice. The physics in this (classical) limit is now clear: For $0 < b < 1$, the energy is minimized by having two of

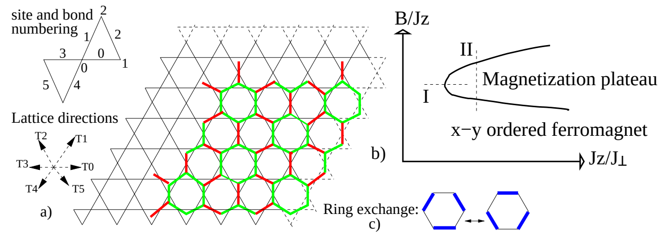

FIG. 1: (color online) (a) Periodic Kagome lattice and the honeycomb net whose bonds pass through the kagome sites. In the plaquette ordered state, red honeycomb edges have no dimer ($\sigma^z = +1$), while green hexagons resonate via the ring-exchange process (shown in (c)). In the alternate columnar state at the same wavevector, dimers cover all red edges ($\sigma^z = -1$) but not green ones. (b) Schematic phase diagram, showing the scans I and II discussed in text.

the spins in each triangle pointing up and one pointing down, which yields a magnetization equal to 1/3 of the saturation magnetization, while for $b > 1$, the ground state magnetization is locked to the saturation value by having all spins pointing up. Thus, one expects a magnetization plateau at 1/3 of the saturation magnetization in the vicinity of $b = 0.5$, where the energy gap to change in magnetization is largest. In this (classical) limit, the ground state has extensive degeneracy, as may be easily seen by noting that the manifold of low-energy configurations can be mapped to the perfect dimer covers of the honeycomb lattice whose edges pass through the kagome sites (with each down spin corresponding to a dimer covering the corresponding honeycomb edge).

Let us now turn on a small $J_\perp$. Apart from an unimportant constant shift in energy, the leading non-trivial effect of this perturbation is easily seen to arise at third order in degenerate perturbation theory and correspond to a 'ring-exchange' term which allows flippable hexagonal plaquettes to resonate with amplitude $t \sim -J_\perp^3/J_z$ (Fig 1 (c)). This quantum dimer model on the honeycomb lattice is known to be in a crystalline 'plaquette' state that breaks the lattice translation symmetries of the honeycomb lattice in order to maximize the number of independently flippable plaquettes from which the system can gain kinetic energy[18]. This implies a ground state with long-range density wave order of the σ^z and of the bond energies $\sigma_i^+\sigma_j^- + h.c.$ (Fig 1).

Thus, the plateau state is stable for large finite $J_z/J_\perp$ and is therefore expected to occupy a lobe in the B/J_z-$J_z/J_\perp$ plane (Fig 1). Clearly, the tip of this lobe represents a special point along the locus of plateau transitions as the vicinity of the tip is distinguished by the presence of low energy 'particle-hole' symmetry corresponding to equal energies for 'quasiparticle' and 'quasihole' excitations (here quasiparticles and quasiholes are distinguished by the sign of the magnetization deviation from 1/3 that they induce by their presence). Given that the plateau state breaks lattice translation symmetry, the transition to the ferromagnet (nematic) at the

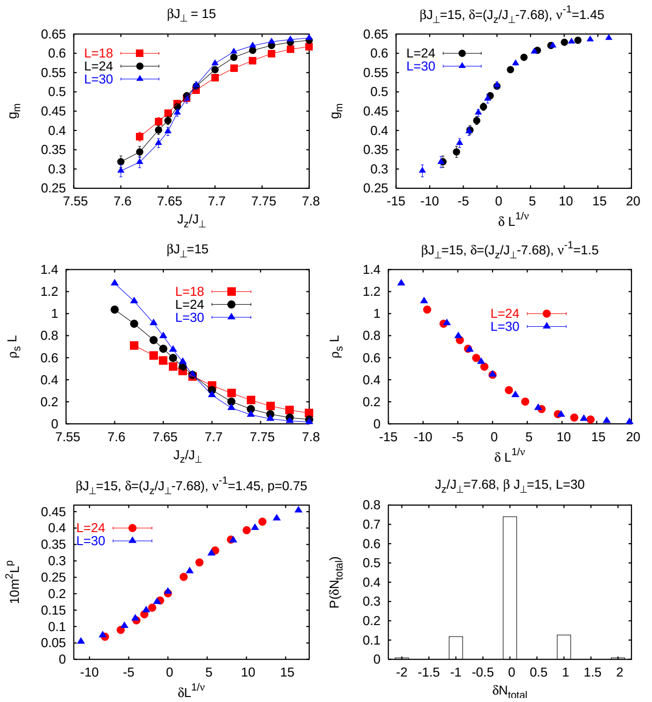

FIG. 2: Numerical evidence for direct second order transition at the particle-hole symmetric tip of magnetization plateau lobe (scan I). Here $m^2 = S^{00}_\rho(Q,\omega_n=0)/4L^2\beta$, while $g_m = 1-\langle m^4\rangle/(3\langle m^2\rangle^2)$ is the standard Binder cumulant of the spin-density wave order parameter and $\delta N_{tot} \equiv 0.5\delta\sigma^z_{tot}$

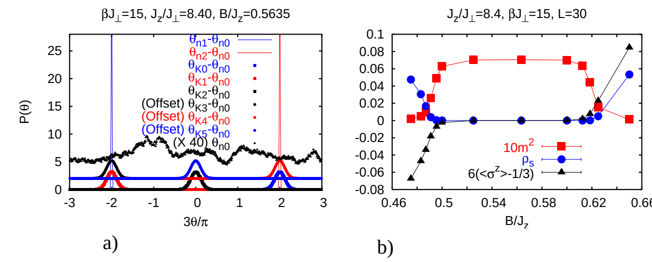

FIG. 3: a) Histograms of relative and absolute phase of all order parameters. b) Vertical scan (II) showing plateau state at $J_z/J_\perp = 8.4$.

tip and away presents interesting possibilities: Conventional Landau theory would predict either an intermediate phase with both orders present or a first order transition. However recent work[19] has shown that Landau theory itself can fail in closely related bosonic models - the result in such cases is expected to be an unusual direct second order phase transition.

Numerical results: The foregoing provides the motivation for our numerical study of the $S = 1/2$ XXZ model at large $J_z/J_\perp$ and finite field $B \lesssim 0.5J_z$. We use the well-documented stochastic series expansion (SSE) QMC method[20] to access the phase diagram. (At large values of $J_z/J_\perp$, some modifications developed recently[13,21] were used to improve the algorithmic efficiency). Most of our data is on $L\times L$ samples (where L is number of unit cells) with periodic boundary conditions and L a multiple of six ranging from 18 to 30 at inverse temperatures β ranging from $5/J_\perp$ to $15/J_\perp$. We use standard SSE estimators[20] to calculate the ferromagnetic stiffness ρ_s, the equal time ($C^{\alpha\alpha'}_\rho(q,\tau=0) = \langle\sigma^z_\alpha(q)\sigma^z_{\alpha'}(-q)\rangle$) and static correlators ($S^{\alpha\alpha'}_\rho(\vec{q},\omega_n=0) = \int_0^\beta d\tau C^{\alpha\alpha'}_\rho(\vec{q},\tau)$) of σ^z_α, and the static correlator of the 'kinetic energy' $K_l = (\sigma^+\sigma^- + h.c.)_l$ on link l ($S^{\alpha\alpha'}_{\mathrm{K}}(\vec{q},\omega_n=0) = \int_0^\beta d\tau C^{\alpha\alpha'}_{\mathrm{K}}(\vec{q},\tau)$) (here α and α' refer to the 3 basis sites and six bond orientations in a unit cell, and all site and bond types shown in Fig 1 are assigned the coordinates of site type 0 when defining the fourier transform).

By analyzing the L and β dependence of the Bragg peaks at $\pm Q = \pm(-2\pi/3, 2\pi/3)$ (components refer to projections along T_0 and T_1 (Fig 1)) seen in the static correlation functions of σ^z and K_l, we conclude that spatial order is established at these wavevectors when ferromagnetism is destroyed in the plateau state; the observed wavevector Q is the ordering wavevector of the plaquette and columnar states of Fig 1. The static structure factors near the onset of the plateau state also reveal the presence of an interesting 'dipolar' structure somewhat analogous to the dipolar part of dimer correlators in the classical honeycomb lattice dimer model[22]. These seem to simply reflect the *local* magnetization 1/3 constraint imposed by the B and J_z terms in this region of parameter space[23] and persist across the transition into the ordered state.

To further probe the nature of the ordering, we also measure the statistics of the phases $\theta_{n\alpha}$, $\theta_{K\alpha}$ of nine complex order parameters $\psi_{n\alpha} = \sigma^z_\alpha(Q,\omega_n=0)$ and $\psi_{K\alpha} = K_\alpha(Q,\omega_n=0)$. From Fig 3 (a), we see that that all relative phases are essentially pinned by the energetics of the plateau state and there is one independent phase degree of freedom which we take to be θ_{n0} ($\theta_{n0} = 0, \pm 2\pi/3$ correspond to the three equivalent plaquette ordered states while $\theta_{n0} = \pi, \pm\pi/3$ correspond to the alternative 'columnar' states at the same wavevector). Clearly, this overall phase is very weakly pinned (compared to the relative phases) even at low temperatures relatively far from the critical region, but the presence of distinct Bragg peaks in the kinetic energy correlator strongly suggest that the ordering is of the plaquette type as simple caricatures of the columnar state contain no dimer resonances.

The statistics of these phases can be interpreted in terms of a Landau free energy F written in terms of the order parameters $\psi_{n\alpha}$, $\psi_{K\alpha}$: $F = F_1(|\psi_{n\alpha}|^2, |\psi_{K\alpha}|^2) + F_2$, where $F_1 = r_1|\psi_{n\alpha}|^2 + r_2|\psi_{K\alpha}|^2 + u_1(|\psi_{n\alpha}|^2)^2 + u_2(|\psi_{K\alpha}|^2)^2 + \ldots$ is insensitive to lattice geometry and phase information and F_2 encodes phase information. When F_1 develops a minimum at non-zero values of its argument, F_2 determines the details of the ordering pattern. The most important terms in F_2 are quadratic invariants which encode the *local* magnetiza-

tion 1/3 constraint on each triangle (imposed by J_z and $B \sim 0.5J_z$), and the kinetic energy gain from having pairs of neighbouring sites exchanging spins ($-J_\perp$). The former may be written as $a(|\Psi_n \cdot I|^2 + |\Psi_n \cdot \Omega^*|^2) - b|\Psi_n \cdot \Omega|^2$ with a and b both expected to be positive on energetic grounds (we use the dot product notation $v \cdot w \equiv \sum_\alpha v_\alpha w_\alpha$, and $\Psi_n \equiv (\psi_{n0}, \psi_{n1}, \psi_{n2})$, $\Omega \equiv (1, e^{2\pi i/3}, e^{4\pi i/3})$, $I = (1,1,1)$), while the latter is given by $c|\Psi_K \cdot \Theta|^2$ with c expected to be negative on energetic grounds (here $\Psi_K \equiv (\psi_{K0}, \psi_{K1}, \psi_{K2}, \psi_{K3}, \psi_{K4}, \psi_{K5})$ and $\Theta \equiv (1, e^{4\pi i/3}, e^{2\pi i/3}, e^{4\pi i/3}, 1, e^{2\pi i/3})$). In addition, symmetries also permit a quadratic cross-term $\left[(\Psi_K \cdot \Theta)(\Psi_n^* \cdot \Omega^*)e^{-2\pi i/3} + h.c.\right]$. Landau theory thus predicts that the relative phases are such that the natural order parameters $\Psi_K \cdot \Theta$ and $\Psi_n \cdot \Omega$ have maximum modulus and relative phase of $2\pi/3$ (assuming the coefficient of the cross-term is negative), and clearly this correctly reproduces the phase relationships exhibited by the data (Fig 3 (a)). Finally, the absolute phase is expected to be chosen by cubic terms of the form $Re\left[(\Psi_n \cdot \Omega)^3\right]$, and our data suggests that the *renormalized* value of the corresponding coefficient is extremely small even away from the phase boundary.

We have also studied the nature of the phase boundary between the plateau state and the ferromagnet by performing several scans, of which we show data for two here. The first scan (scan I shown in Fig 1) at constant $B/J_z = 0.5635$ was chosen as it intersects the phase boundary at a particle hole symmetric point which we identify as the tip of the plateau lobe. The sharpness of the crossings seen in the plots (Fig 2) of the Binder cumulant g_m and of $\rho_s L$ for different sizes strongly suggest that we have reached the asymptotic low temperature regime and provide indications that the transition is a direct second order transition at $J_z/J_\perp \approx 7.68 \pm 0.02$ with $z = 1$. From a scaling collapse of these crossing curves, we estimate $1/\nu \approx 1.45 \pm 0.2$, while similar analysis for the order parameter m^2 gives $2\beta/\nu \approx 0.75 \pm 0.1$; however, we emphasize that these are *estimates* and a more detailed study with much larger sizes is needed to definitively rule out a very weak first order jump or small coexistence region and obtain precision values of exponents. The second, vertical scan (scan II) was performed at $J_z/J_\perp = 8.4$ primarily to confirm the existence of the plateau state over an appreciable range of B, and yields a plateau state for $0.49 \lesssim B/J_z \lesssim 0.62$ (Fig 3 (b)). Near the transition points, no clear evidence of a first order jump is seen, nor is there a well-resolved region with coexisting order parameters (further details regarding the nature of these transitions will be discussed separately[23]).

Summary: To summarize, we have demonstrated that $S = 1$ kagome antiferromagnets with moderately strong single-ion anisotropy of the easy-axis type exhibit an interesting spin-nematic state at and in the vicinity of $B = 0$, as well as a lattice-symmetry broken spin-density wave magnetization plateau state at 1/3 magnetization for $B \sim 0.5J$. We have also presented numerical evidence that the transition between these is of an unusual direct second order type at least at the tip of the plateau lobe in the B/J-D/J plane. We hope our work will provide some impetus to look for these effects in anisotropic $S = 1$ kagome magnets.

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